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Remarks

Claims 2, 3 and 5 to 12 are currently pending in this case. Claims 7 to 11 stand withdrawn and Claims 2, 3, 5, 6 and 12 stand rejected.

Applicants respectfully request that the Examiner enter and consider the changes made in Claim 12 which are indicated in the Listing of Claims set forth in Appendix I attached to this paper. The changes merely serve to further emphasize that the wording "in powder form for use as a fluidized-bed catalyst" in Claim 12 is an essential characteristic of applicants' catalyst and is, therefore, more than a mere indication of the intended use. The revision of the wording of Claim 12 does not add new matter. Moreover, the changes made in the wording of Claim 12 are merely editorial in nature. The revision of Claim 12 therefore cannot be deemed to necessitate an extended search and/or examination. Entry of Claim 12 as herewith submitted and consideration of revised Claim 12 is therefore deemed equitable. Favorable action is solicited.

The Examiner has maintained the rejection of applicants' Claims 2, 3, 5, 6 and 12 under 35 U.S.C. §102(b) as being anticipated by the teaching of *Courty et al.* (US 4,381,415) and by the teaching of *Convers et al.* (US 4,460,699). In this context the Examiner takes the position that applicants' Claim 12 differs from Claim 1 which it replaces merely in the indication of an intended use of the catalyst. Applicants respectfully disagree.

As specifically provided in Claim 12, applicants' catalyst is "in powder form for use as a fluidized-bed catalyst" (emphasis added). All words in a claim must be considered in judging the patentability of a claim against the prior art1). Moreover, anticipation under Section 102 can be found only if a reference shows exactly what is claimed2), and the test for anticipation is one of identity, cf. the identical invention must be shown in the reference in as complete detail as is contained in the claim3). Neither the teaching of Courty et al. nor the teaching of Convers et al. discloses a catalyst which comprises

¹⁾ In re Wilson, 424 F.2d 1382, 165 USPQ 494 (CCPA 1970).

²⁾ For example <u>Titanium Metals Corp. v. Banner</u>, 778 F.2d 775, 227 USPQ 773 (CAFC 1985); <u>In re Marshall</u> 577 F.2d 301, 198 USPQ 344 (CCPA 1978); <u>In re Kalm</u> 378 F.2d 959, 154 USPQ 10 (CCPA 1967).

³⁾ For example <u>Richardson v. Suzuki Motor Co.</u>, 868 F.2d 1226, 9 USPQ2d 1913 (CAFC 1989)

at least one active component and a catalyst support comprising δ -Al₂O₃ in an amount detectable by X-ray diffractometry which is in powder form. Even without the recitation of the intended use of applicants' catalyst, there is accordingly a clear structural distinction between applicants' catalyst and the catalysts which are disclosed by the references applied by the Examiner.

The Examiner remarks that a claim is met where a prior art structure is capable of performing the intended use. The respective statement is, however, only applicable in situations where the intended use is the only distinguishing feature. Here, the claimed subject matter is distinguished over the prior art structure by the requirement that the catalyst be in powder form, and the Examiner's remark does not apply. Concerning the Examiner's remark that a process of making must provide for a manipulative difference from the prior art process it is respectfully noted that applicants' process requires "impregnating pulverulent δ -Al₂O₃-containing support" (emphasis added). The manipulative difference between applicants' process and the prior art processes, accordingly, resides in the fact that a pulverulent support material is employed. It is therefore respectfully requested that the rejections under Section 102(b) be withdrawn. Favorable action is solicited.

For completeness sake it is further respectfully submitted that the teachings of *Courty et al.* and of *Convers et al.*—when taken alone or in combination with one another, or when one is taken in view of the other— are insufficient to render applicants' invention *prima facie* obvious within the meaning of 35 U.S.C. §103(a). Both references relate to catalysts for fixed bed systems⁴). It is well known in the art and briefly addressed by *Convers et al.* that particles which are smaller than a particular minimum size are unsuitable as fixed-bed catalysts⁵). It is, correspondingly, well known that catalyst powders are unsuitable for fixed bed systems.

In order to render a claimed invention obvious within the meaning of Section 103(a), the prior art has to provide for a teaching or suggestion to make the claimed combination and for a reasonable ex-

⁴⁾ Concerning the teaching of Convers et al. note, for example, the title and the abstract of US 4,460,699. Concerning the teaching of Courty et al., note the marked parts in the enclosed copy of Ullmann's Encyclopedia of Industrial Chemistry, Vol. Al3, p. 496 (5th Edition, VCH Verlagsgesellschaft mbH, 1989) on "Catalytic Reforming of Hydrocarbons".

⁵⁾ Note col. 3, indicated line 62, to col. 4, indicated line 1, of US 4,460,699.

pectation of success⁶⁾, and the modification of the prior art which is necessary to arrive at the claimed invention cannot render the prior art invention which is being modified unsatisfactory for its intended purpose⁷⁾. Since catalysts in powder form are unsuitable as fixed-bed catalysts, a transformation of the catalyst particles which are taught by *Courty et al.* and/or *Convers et al.* into a powder form would render those catalysts unsuitable for the purposes of the inventions disclosed in the prior art references. Applicants' invention is, therefore, not rendered *prima facie* obvious by the teaching of *Courty et al.* and/or *Convers et al.*

In light of the foregoing, the application should be in condition for allowance. Early action by the Examiner would be greatly appreciated.

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Respectfully submitted,

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Encl.: THE LISTING OF CLAIMS (Appendix I)

Ullmann's Encyclopedia of Industrial Chemistry, Vol. A13, pp. 496-497

(5th Edition, VCH Verlagsgesellschaft mbH, 1989)

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^{6) &}lt;u>In re Vaeck</u>, 947 F.2d 488, 20 USPQ2d 1438, 1442 (CAFC 1991)

⁷⁾ If proposed modification would render the prior art invention being modified unsatisfactory for its intended purpose, then there is no suggestion or motivation to make the proposed modification (<u>In re Gordon</u>, 733 F.2d 900, 221 USPQ 1125 (CAFC 1984)).

APPENDIX I:

THE LISTING OF CLAIMS:

- 1. (canceled)
- 2. (previously presented) A catalyst as in claim 12, wherein the catalyst support comprises from 10 to 100% by weight of δ -Al₂O₃.
- 3. (previously presented) A catalyst as in claim 12, wherein the active components employed are from 1 to 15% by weight of copper, from 0.1 to 6% by weight of alkali metals, and from 0 to 5% by weight of alkaline earth metals, rare-earth metals or mixtures thereof.
- 4. (canceled)
- 5. (previously presented) A process for the preparation of the catalyst of claim 12, which comprises impregnating pulverulent $\delta-\text{Al}_2\text{O}_3-\text{containing support}$ with salts of copper, alkali metals and, optionally, alkaline earth metals, rare-earth metals or mixtures thereof, separately from one another or together, optionally with the addition of acids or oxidants.
- 6. (previously presented) The process of claim 5, wherein the salts employed are chlorides.
- 7. (withdrawn) The use of a catalyst for heterogeneously catalyzed reactions as claimed in claim 1 for exothermic gas-phase reactions.
- 8. (withdrawn) The use of a catalyst for heterogeneously catalyzed reactions as claimed in claim 1 for oxychlorination reactions.
- 9. (withdrawn) The use of a catalyst for heterogeneously catalyzed reactions as claimed in claim 1 for the oxychlorination of ethylene to 1,2-dichloroethane.
- 10. (withdrawn) A process for the preparation of 1,2-dichloroethane, which comprises reacting ethylene with hydrogen chloride and air or oxygen in the presence of a catalyst as claimed in claim 1 at a temperature of from 150 to 400°C and a pressure of from 1 to 10 bar.
- 11. (withdrawn) The use of a catalyst for heterogeneously catalyzed reactions as claimed in claim 1 for partial oxidation reactions.

12. (currently amended) An oxychlorination catalyst which is in powder form for use as a fluidized-bed catalyst, and which comprises at least one active component with a catalyst support comprising $\delta\text{-Al}_2\text{O}_3$ in an amount detectable by X-ray diffractometry.

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Ullmann's Encyclopedia of Industrial Chemistry

Fifth, Completely Revised Edition

Volume A 13:

High-Performance Fibers to Imidazole and Derivatives

Editors: Barbara Elvers, Stephen Hawkins,

Michael Ravenscroft, Gail Schulz



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Production Director: Maximilian Montkowski Production Manager: Myriam Nothacker

Library of Congress Card No. 84-25-829

Deutsche Bibliothek, Cataloguing-in-Publication Data:

Ullmann's encyclopedia of industrial chemistry / ed.: Barbara Elvers... [Ed. advisory board Hans-Jürgen Arpe...]. — Weinheim; Basel (Switzerland); Cambridge; New York, NY: VCH

Teilw. executive ed.: Wolfgang Gerhartz. —

Teilw. mit d. Erscheinungsorten Weinheim, Deerfield Beach, Fl. —

Teilw. mit d. Erscheinungsorten Weinheim, New York, NY

Bis 4. Aufl. u. d. T.: Ullmanns Encyklopädie der technischen Chemie

NE: Gerhartz, Wolfgang [Hrsg.]; Elvers, Barbara [Hrsg.]; Encyclopedia of industrial chemistry

Vol. A. Alphabetically arranged articles.

13. High-Performance Fibers to Imidazole and Derivatives. — 5th, completely rev. ed. — 1989

ISBN 3-527-20113-0 (Weinheim);

ISBN 0-89573-163-0 (New York)

British Library Cataloguing in Publication Data

Ullmann's encyclopedia of industrial chemistry.

Vol. A 13, High-Performance Fibers to Imidazole and Derivatives

1. Industrial chemistry

I. Elvers, Barbara II. Rounsaville, James F. III. Schulz, Gail

661

ISBN 3-527-20113-0

© VCH Verlagsgesellschaft mbH, D-6940 Weinheim (Federal Republic of Germany), 1989.

Distribution

VCH Verlagsgesellschaft, P.O. Box 1011 61. D-6940 Weinheim (Federal Republic of Germany)

Switzerland: VCH Verlags-AG, P.O. Box, CH-4020 Basel (Switzerland)

Great Britain and Ireland: VCH Publishers (UK) Ltd., 8 Wellington Court, Wellington Street, Cambridge CB1 1HW (Great Britain)

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Cover design: Wolfgang Schmidt

Composition, printing, and bookbinding: Graphischer Betrieb Konrad Triltsch, D-8700 Würzburg Printed in the Federal Republic of Germany

cally possible at a temperature below that of thermal decomposition. Propane, butane, and alkanes in the detergent alkylate range (ca. $C_{10} - C_{14}$) have all been catalytically dehydrogenated on a large scale. Little advantage is gained by catalysis in the dehydrogenation of C_2 hydrocarbons, because the equilibrium is unfavorable at a temperature below that required for thermal decomposition.

Cracking (cleavage of the carbon-carbon bond) is energetically favored over cleavage of the carbon-hydrogen bond, and catalysts such as nickel, which are excellent for hydrogenation of alkenes, lead to extensive formation of methane when applied to dehydrogenation.

The reaction expressing the general alkane – alkene equilibrium is

$$C_nH_{2n+2} \rightleftharpoons C_nH_{2n} + H_2$$

Alkenes are favored by lower pressure, as expressed by the equation

$$C^2/(1-C^2)=K/p$$

where C is the fraction dehydrogenated, K is the equilibrium constant, and p the pressure in atmospheres. This equation holds for the conversion of alkanes to monoenes and of monoenes to dienes. Conversion of alkanes to dienes (e.g., butane to butadiene)

$$C_nH_{2n+2} \iff C_nH_{2n-2} + 2 H_2$$

is related to pressure by the equation

$$4C^3/(1-C)(1+2C)^2 = K/p^2$$

Conversion is increased by operation at lower pressure or by use of a diluent.

Butenes and butadiene are produced by dehydrogenation of butane, although the process finds increasing competition from oxidative dehydrogenation and from thermal cracking (→ Butadiene, A4, pp. 435–440; → Butenes, A4, p. 487). Operating conditions vary: the temperature is 550–650 °C and the pressure 30–300 kPa, contact times are a few seconds. Catalysts coke rapidly and are oxidatively regenerated every 10–100 min. By mixing the catalyst with inert material, much of the heat liberated during oxidation can be captured and used to drive the endothermic dehydrogenation. Most catalysts in this service are based on chromia–alumina. These catalysts are strongly inhibited by water

vapor; the butane feed must, therefore, be dried carefully.

Other catalysts, such as nickel-calcium phosphate stabilized with chromia, iron oxide promoted by potassium, and chromium oxides, require the presence of steam to help keep the catalyst in the proper oxidation state. Excessive coking occurs if these catalysts become reduced. Steam also acts as a source of heat and shifts the conversion toward the dehydrogenated product by lowering the partial pressure of the latter.

2.3. Catalytic Reforming of Hydrocarbons

The objective of catalytic reforming is the conversion of saturated hydrocarbons to aromatics as selectively as possible. This process is the chief source of high-octane fuel and of aromatic hydrocarbons. Catalytic reforming involves a complex series of reactions in which dehydrogenation figures prominently. Catalysts are dual-functional, having both a metallic element to catalyze dehydrogenation-hydrogenation and an acidic function to catalyze hydrocarbon rearrangements. Catalysts typically contain a small amount of highly dispersed platinum (<1%) supported on alumina $(150-300 \text{ m}^2/\text{g})$, as well as a second metal such as rhenium. Polymetallic catalysts permit operation under more severe conditions; they also possess improved stability and activity.

Typical operating conditions are 430-530 °C and 1.0-3.5 MPa. A unit consists of several, frequently four, fixed-bed reactors in series. Heated naphtha enters the first reactor, where the major reaction is endothermic dehydrogenation of cycloalkanes accompanied by a decrease in temperature. The effluent is reheated and the process continued for all reactors. The liquid product contains 60-70 wt \% aromatics; the gaseous product contains light hydrocarbons admixed with 60-90 mol\% hydrogen. The gaseous portion is recycled to the first reactor in a ratio of 5-10 mol of recycle gas per mole of naphtha to maintain a high partial pressure of hydrogen in the system. The presence of hydrogen diminishes the yield of aromatics, but hydrogen is necessary to maintain catalyst activity by retarding formation of hydrocarbon residues that poison the catalyst. An aim of processing is operation at as low a partial pressure of hydrogen as possible, commensurate with good catalyst life.

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Dehydrogenation of cyclohexanes is the most facile of all reactions that occur in reforming. On the other hand, only traces of olefins can be formed by dehydrogenation of alkanes under reforming conditions, because of an unfavorable equilibrium. Nonetheless, dehydrogenation is of considerable importance because olefins appear to be intermediates in the overall process [22].

2.4. Liquid-Phase Dehydrogenation

The constraints imposed on dehydrogenation by equilibrium considerations can be loosened if hydrogen is removed from the products as they are formed. Dehydrogenation in the liquid phase in an open vessel fulfills this goal. Hydrogen escapes from the system, whereas products and reactants remain in the vessel or are returned after condensation. The lower temperature limit is then set not by the equilibrium but by a convenient reaction rate.

In practice, the substrate, neat or dissolved in an inert solvent of convenient boiling point, is refluxed in the presence of catalyst. Hydrogen escapes while other volatile products are condensed and returned, with or without removal of the product. If the product is removed, the operation can be made continuous.

All the considerations involving the influence of mass transport on the rate of reaction (see Section 1.1) apply in reverse to dehydrogenation. Good stirring helps to remove liberated hydrogen from the vicinity of the catalyst, and reflux facilitates hydrogen removal from the liquid. If the substrate or product is thermally unstable at normal reflux temperature, the temperature can be lowered by reducing the pressure. Liquid-phase dehydrogenation is used in the production of a number of fine chemicals of limited thermal stability.

Dehydrogenation may be facilitated by the use of a hydrogen acceptor to remove hydrogen from the system as it is formed [23].

$$\begin{array}{c}
NH \\
CN \\
+ \\
N \\
N
\end{array}$$

$$\begin{array}{c}
170^{\circ}C \\
20 \text{ kPa}
\end{array}$$

$$\begin{array}{c}
NH_{2} \\
CN \\
+ \\
N
\end{array}$$

$$\begin{array}{c}
NH_{2} \\
+ \\
N
\end{array}$$

The hydrogen transfer reaction above may be written generally as follows:

$$AH_2 + B \longrightarrow A + BH_2$$

These hydrogenation—dehydrogenation reactions are useful for carrying out hydrogenations in places where hydrogen is unavailable or its use is not permitted; only simple equipment is required [24]. Hydrogen transfer reactions often give better yields of hydrogenated products than are obtained by direct hydrogenation.

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Hydrometallurgy → Metallurgy